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Antiferroelectrics for energy storage applications: A Review

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Abstract: The requirement of electrical energy to power modern equipment and electronics is increasing day by day. This leads to the need for developing methods for generation and storage of electrical power which are highly efficient. Hence, a lot of research work is focused on the development of efficient energy storage materials. So that they can be utilized for various industrial applications. Modern electronic and electrical devices can be revolutionized by using dielectric based solid state capacitors. Among the popular dielectric materials, antiferroelectrics display higher energy density and higher power density as compared to their linear dielectric and ferroelectric counterparts. They also possess low dielectric loss, low remnant polarization, low coercive field, higher material efficiency and fast rate of discharging. Due to all these characteristics, antiferroelectric materials have a huge potential to be used in various energy storage applications. This review paper presents basic facts about antiferroelectric materials regarding their definition and energy storage behavior.

Keywords: Antiferroelectric, energy storage materials, BNT, dielectric

Introduction: Fossil fuel resources are depleting at a fast rate and in addition they are also responsible for causing a lot of air pollution and global warming. This leads to the requirement of various renewable and clean sources of energy. [1- 4] Conversion of this energy to electricity is very nice option in association with the development of reliable and efficient electrical energy storage devices needed for proper implementation of electricity that is being generated from these renewable sources of energy. [1,5-9] It is very important to store electrical energy properly because electricity is the primary need to power much of our available technology now a days. Hence, a lot of research work is being done in order to develop some suitable materials that should possess enhanced energy and power density so that requirements of modern electric devices and machines can be met properly. [10] Majority of the electrical energy storage devices available today include electrochemical capacitors, batteries and dielectric capacitors. Of these, electrochemical capacitors have medium energy density and power density. Also, they suffer from low operating voltage, large leakage current and high cost than dielectric capacitors. [11,12]

Batteries possess high power density but they do not provide desired power output. While reverse is true in case of dielectric capacitors as they have lower energy density but exhibit high power output. [13,14] This difference is due to their different mode to store energy . In case of batteries, the electrical energy is first converted into chemical energy and then again reconverted in electrical energy using electrochemical cell. [15,16] So, chemical reaction rate will determine rate of these conversions and release of energy again in case of batteries. While dielectric capacitors store energy in form of localized electric fields or dipole moments existing in the dielectric material due to its polarization through electrical energy. [13,14] This energy is stored over several charging cycles and then it can be released at a rapid rate in order to produce electrical pulses of known power. Thus, these capacitors can be used in many devices that need high power electric supply. [13-18]. For capacitor applications, the maximum energy storage density per unit volume, U for a dielectric material is given by the equation

$$U = \frac{1}{2}\varepsilon_0 \varepsilon_r \varepsilon_b^2$$

Here, ε_0 is dielectric permittivity of free space, ε_r is relative permittivity of material and E_b is the dielectric breakdown electric field of material.[19]

This equation shows that energy density of material increases with the increase in dielectric constant and dielectric breakdown strength of material. [19]. Amongst dielectrics, we have linear dielectrics, ferroelectrics, relaxor ferroelectrics and antiferroelectrics that can be used for making electrical capacitors. Linear dielectrics have low value of dielectric constant and they have high dielectric breakdown strength. While ferroelectrics are characterised by large value of dielectric constant but they have lower value of dielectric breakdown strength and large dielectric losses. So, these two are unsuitable for use in applications associated with storage of energy. Relaxor ferroelectrics with almost zero remnant polarization are good option for capacitor applications but the materials with relaxor characteristics are relatively rare. [20] Antiferroelectric materials have a unique feature of phase transition in ferroelectric - antiferroelectric state induced by electric field and because of this, these materials possess higher energy storage density, low remnant polarization, fast rate of discharging and low coercive field. [21,22] AFE Phase stability, maximum polarization of induced ferroelectric phase and breakdown strength of the material need to be enhanced in order to increase the energy storage property of AFE ceramics.

Definition of antiferroelectrics - Antiferroelectrics are typically made up of two sublattices with antiparallel polarization directions. These antiparallel polarizations are equal in magnitude but opposite in direction and thus results into a net zero macroscopic polarization. [23-25] Antiferroelectrics are usually characterized by double hysteresis loop i.e. Polarization – electric field (P - E) loop. In antiferroelectrics, there occurs a phase transition between antiferroelectric and ferroelectric states induced by an external electric field. As electric field is increased beyond certain strength then AFE to FE phase transition takes place at electric field E_F, known as critical forward phase switching field. When we decrease the electric field then FE state goes back to AFE state i.e. FE - AFE phase switching takes place at E_A, known as critical backward phase switching field. This field induced phase transition also leads to change in unit cell volume which further results in field induced strain produced in the material. Values of these phase switching fields E_F and E_A are also affected by change of temperature and pressure.

Perovskite structure of antiferroelectrics and tolerance factor - Most of the antiferroelectrics are perovskites that can be represented by ABO_3. This structure is composed of corner linked oxygen octahedra having B-site cation at the center and A-site cation present at the interstice. Tolerance factor (t) given by Goldschmidt is used to determine the stability of these perovskite ABO_3 structures and it is given by the following expression [27]

$$t = \frac{R_A + R_0}{\sqrt{2}(R_B + R_0)}$$

Here, R_0, R_A *and* R_B are the radii of oxygen ions, A-site cations and B-site cations respectively. Value of tolerance factor $t > 1$ for ferroelectrics and $t < 1$ for antiferroelectrics [27]. Thus, lower tolerance factor will lead to more stable AFE phase.

Energy Storage in Antiferroelectrics -

To calculate the recoverable energy storage density (W_r) of dielectric material, we have the following equation [26].

$$W_r = \int_{P_r}^{P_{max}} E dP$$

Here, E is applied electric field and P_r and P_{max} are respectively the remnant polarization and maximum polarization.

The energy efficiency η can be written as

$$\eta = W_r/(W_r + W_{loss})$$

Where W_{loss} represents the loss of energy density [26]. Recoverable energy density is given by the area between P-E curve and polarization axis while W_{loss} is represented by the area within P-E loop. Antiferroelectrics with special feature of transition between antiferroelectric and ferroelectric phases induced by electric field produce negligible P_r and large P_{max} and serve as good alternatives for utilization in enhanced energy storage over linear dielectrics and ferroelectrics. In addition, the material is also required to have large values of critical phase switching fields E_F and E_A as well as high breakdown strength.

The antiferroelectric ceramics that have been developed for energy storage applications so far include both lead containing compounds like $PbZrO_3$, $PbHfO_3$, PLZST, PNZST as well as lead free ceramics such as silver niobate based, sodium niobate based and BNT based systems. In $(Pb_{0.94} La_{0.04}) (Zr_{0.49} Sn_{0.5} Ti_{0.01}) O_3$ ceramics, a very high 9.6J/cm^3 recoverable energy density with high efficiency 90.2% was found by Xihong Hao et al [28].

Although lead based AFEs show very good energy performance but due to increasing concern towards environment, it is necessary to phase out lead from various AFE ceramics because of its toxic nature. Therefore, a lot of research work has been done in past few years to develop lead free antiferroelectrics like silver niobate based, sodium niobate based and BNT based systems [29]. The energy performance of lead free antiferroelectris is poor with efficiency less than 80% in comparison with lead based antiferroelectrics [29]. These low values of energy density and efficiency of lead free AFEs cause hindrance in their real application for various energy storage devices.

Disadvantages associated with antiferroelectric materials- Although there are many advantages of using AFEs in energy storage devices but there are few drawbacks also which put a limit on their practical applications. First factor is piezoelectric noise that is produced in the structure due to strain induced by electric field when it increases beyond a certain threshold value and AFE to FE phase transformation takes place. This piezoelectric noise has to be reduced otherwise it may damage the structure altogether. [30] Second factor that affects the energy performance of AFEs is polarization fatigue. In this, saturation polarization lowers on enhancing continuation of electric cycles. It can result into higher dielectric loss, higher remnant polarization and lower dielectric constant. [30] If we could overcome these two disadvantages, then AFEs would perform better for storage of energy

Conclusion – The energy storage performance of lead containing antiferroelectrics are superior as compared to that of lead free antiferroelectrics. Therefore, it is impossible yet to replace entirely the lead based AFEs by lead free AFEs. Amongst lead free AFE systems, $AgNbO_3$ based, $NaNbO_3$ based and BNT based AFEs prove to be the potential candidates for energy storage applications. A lot of research work is being carried out on tuning the composition of environment friendly antiferroelectrics i.e. the AFEs without lead to have a control on various important characteristics like critical phase switching fields, phase transition between AFE - FE phases induced by electric field and strain response. Sincere efforts should be put to develop new lead free AFE materials that can match the level of performance of best available lead based antiferroelectric for energy storage applications.

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